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OBLON SPIVAK ETC



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**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

IN RE APPLICATION OF:

Laura ZANIBELLI et al

SERIAL NO.: 09/539,877

FILED: March 31, 2000

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: GROUP ART UNIT: 1764

: EXAMINER: T. Nguyen

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FOR: CATALYTIC COMPOSITION FOR THE  
UPGRADING OF HYDROCARBONS HAVING  
BOILING POINTS WITHIN THE NAPHTHA RANGE

**DECLARATION UNDER 37 C.F.R. §1.132**

ASSISTANT COMMISSIONER FOR PATENTS  
WASHINGTON, D.C. 20231

SIR:

Now comes Marco Ferrari who deposes and says that:

- 1) I am one of the joint inventors of the present application.
- 2) In 1984 I received a degree in chemistry from the Università degli Studi, Milan. *M. Ferrari*
- 3) Since 1990 I have been employed by Enitecnologie S.p.A where I have been engaged as a researcher in the study of catalysis.
- 4) I have read the specification of the present application.
- 5) I have read the outstanding Office Action from the United States Patent Office and the references cited and applied therein.
- 6) In order to demonstrate the superior ability of the catalyst of the hydrodesulfurization process of the invention to conduct isomerization of a hydrocarbon material while minimizing hydrogenation of olefins in the material, the following comparative experiment has been conducted.

### Comparative Experiment A

#### Preparation of Catalyst

A catalyst outside the scope of the catalyst employed in the process of the present invention is prepared as described in EP 748652. A 1.04 g amount of Co nitrate is dissolved in 47.16 g of BuOH, at room temperature. 1.03 g of  $\text{Si}(\text{OC}_2\text{H}_5)_4$  (tetraethylorthosilicate) and 26.53 g of  $\text{Al}(\text{OC}_4\text{H}_9)_3$  (aluminum sec-butoxide) are added to the alcohol solution and the mixture is heated to 60° C for 10 min, thereby obtaining suspension C1.

A 1.47 g amount of EMA is dissolved in 17.56 g of TPAOH (solution at 19,2 %), at room temperature, thereby obtaining solution C2, (pH=11).

Solution C2 is slowly poured into the suspension C1 while heating and with stirring, thereby obtaining a highly viscous fluid which is maintained at 60° C for 1 hour. This is followed by ageing the fluid at room temperature for 21 hours, drying the material in a vacuum oven at 100° C for 6 hours, and calcining the dried material in a muffle furnace under the following temperature regime: heating to 200° C (5° C/min); pausing at 200° C for 2 hours; heating to 550° C (5° C/min); pausing at 550° C for 3 hours and spontaneously cooling to room temperature. The characteristics of the material obtained are shown in Table 1.

TABLE 1

Co (wt %)	Mo (wt %)	Co/Mo (mol)	$A_{\text{surf}}$ (m <sup>2</sup> /g)	$V_{\text{pores}}$ (cm <sup>3</sup> /g)
2.3	8.9	0.42	360	0.74

#### Catalytic Activity

The results of catalytic action using the catalyst above in the treatment of a model charge that is representative of the composition of an FCC gasoline in terms of content and olefinic cut,

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set forth below:

- 30 % by weight of 1-pentene;
- 0.25 % by weight of thiophene (1000 ppm S);
- the balance to 100 is n-hexane.

Catalyst samples are all activate by the same procedure in a stream of  $H_2S/H_2$ .

Catalytic activity is evaluated using the following parameters:

- a) Hydrodesulfurization conversion (HDS %) calculated as follows:

$$HDS \% = 100 \times (\text{ppm } S_{in} - \text{ppm } S_{out}) / \text{ppm } S_{in}$$

- b) Isomerizing properties ISO % calculated as follows:

$$ISO \% = 100 \times (i\text{-pentanes} + i\text{-pentenes}) / \sum C_i$$

- c) Hydrogenating property HYD % calculated as follows:

$$HYD \% = 100 \times (n\text{-pentane}_{out} / 1\text{-pentene}_{in})$$

Two grams of the above prepared catalyst, diluted with corundum, are charged to a reactor (40-70 mesh) and activated in the presence of  $H_2S/H_2$  (10 % vol) up to 400° C for 3 hours. The system is then brought under hydrogen pressure up to 10 bars and the model feed is sent, with a ratio  $H_2$ /hydrocarbon charge equal to 300 NI/1. The operating conditions and catalytic results are shown in Table 2.

Temp (°C)	WHSV (hrs <sup>-1</sup> )	HDS (%)	ISO (%)	HDS/HYD	HYD/ISO
254	6.6	91.0	2.5	1.2	29.9
282	6.6	92.7	2.5	0.9	40.4

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7) The results in Table 2 show that for a catalyst system based on silica and alumina, but not the same as that which is employed in the process of the present invention, lower HDS/HYD ratios, because of higher HYD values, are obtained. The same is also indicated by the relatively high HYD/ISO ratios obtained indicating significant hydrogenation of olefins and a low extent of isomerization of olefins.

8) The undersigned petitioners declare further that all statements made herein of their own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

9) Further, deponent saith not.

Date: \_\_\_\_\_

Morsten